Carrier dynamics in quantum dot structures with different interdot spacings

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Abstract

Photoexcited carrier dynamics has been examined by time-resolved photoluminescence in InGaAs/GaAs quantum dot structures with dot density ranging from 3.7×10^{8} to 2.4×10^{10} cm^{-2}. The time of carrier transfer into a dot decreases with increasing dot density as well as with photoexcited carrier density and temperature. It is suggested that potential barriers at the barrier, wetting layer and quantum dot interfaces hinder the carrier capture in low-density quantum dot structures.

Keywords: quantum dots, carrier dynamics, photoluminescence

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Carrier dynamics is one of the most researched subjects in the quantum dot (QD) physics because of its fundamental interest and importance for applications. However, most of the studies performed so far have concentrated on effects occurring on a rather long time scale, typically 100 ps to several ns. Experimental studies of fast effects, such as carrier capture, are more scarce and inconclusive. Moreover, most of the studies of carrier dynamics performed so far were carried out on single samples making it difficult to draw general trends about the dependence of carrier dynamics on such QD parameters as, size, confining potential, level structure or QD density.

Here, by means of time-resolved photoluminescence (PL), we study changes inflicted on carrier dynamics in QD structures by the QD density, which in the studied set of samples changes by nearly two orders of magnitude. We find that the QD density has a significant influence on the QD level structure and carrier transfer properties.

The InGaAs/GaAs QD samples were grown by MOCVD using a procedure described in [1]. Different QD densities were obtained by slight variations in substrate miscut angle (θ_m) in GaAs (001), nominally: 0.25, 0.75, 1.25 and 2° towards (110) for the samples A, B, C and D. The miscut angles provide different concentrations of steps, which act as energetically favourable sites for island nucleation on the growth surface. The QD densities for the samples A, B, C and D were equal to 3.7×10^8, 7.0×10^8, 2.6×10^9, 7.3×10^9 cm^{-2}, respectively. High surface density (2.4×10^{10} cm^{-2}) randomly distributed QDs (sample E) were also grown ($\theta_m = 0.05$) at the same temperature (550 °C) but under values of arsine partial pressure to maximise island surface coverage. All the QD structures were capped with a 100 nm thick GaAs cap layer. The dot diameters in all the samples were similar, 25±5 nm. Structural characterisation of the islands was done using Atomic Force Microscopy and plan view Transmission Electron Microscopy (TEM) [2]. In the samples B, C and D, the dots align themselves in multi-island strings at multiatomic step edges.

Time-resolved PL measurements were performed in the temperature range 78 to 300 K using a pulsed Ti:sapphire laser (780 nm, 80 fs, 96 MHz) for excitation. For the PL detection, an upconversion set-up (150 fs temporal resolution) and a synchroscan streak camera, combined with a 0.25 m spectrometer (15 ps temporal resolution) were used. The average excitation intensity was in the range from 0.01 to 10 mW, which corresponds to approximately 2×10^{9} to 2×10^{12} electron hole pairs per square centimetre.

Fig. 1 shows 78 K PL spectra of several QD samples at different times after excitation. There are several changes in the QD spectra, which occur with increasing QD density. The QD ground state emission progressively blue shifts, energy levels become spaced more closely, and the relative intensity of the wetting layer (WL) PL decreases. Smaller intersublevel energy transitions and greater inhomogeneous broadening (FWHM) makes excited states observation more difficult until, for the sample with the highest dot concentration, only one PL peak is observed. The progressive blue shift of the QD energy levels with increasing coverage has been also observed in cw PL experiments and has been attributed to increasing lateral strain fields from the adjacent QDs [2,3]. It will be discussed below that the overlapping strain fields affect carrier dynamics in the QD structures as well.

As can be seen from the spectra, the PL decays at different rates for various QD transitions. These times decrease from 1.5 – 2.5 ps for the ground state to 0.6 ps for the highest excited states. This observation is consistent with previous findings of decreasing PL decay times with increasing number of a QD level [4]. In addition, there is a general trend of smaller carrier lifetimes in structures with larger QD density. We have no reason to assume that the high-density samples contain additional nonradiative recombination channels, as all the QD structures were grown in the same run. Thus, we relate the shorter recombination times in the high-density QD samples to shallower QD potentials. A similar decrease of the

carrier lifetimes has been observed previously when the QD potential was decreased by intermixing [5].

Carrier capture into the QDs has been monitored by measuring PL rise times at the energies of various QD transitions. The measurements were performed at different excitation intensities and temperatures (Figs 2 and 3). With increasing excitation power and temperature, the PL rise times decrease until they reach some asymptotic value of about 2 – 3 ps. The variation of the PL rise time is much more pronounced in the small QD density samples. Figures 2 and 3 present PL rise times for the ground state transitions, however, the PL rise times for different QD levels in the low-density samples coincide within the curve fitting errors of 0.5-1.5 ps indicating ultrafast carrier relaxation in the QDs [6]. On the other hand, close values and similar temperature and excitation intensity dependence of the PL rise times for the WL and the QDs in structures B and C suggest that it is the same effect that limits the carrier transfer into the dots and the WL in these samples. Several effects, such as carrier transport in the GaAs barriers and carrier capture into the QD/WL should be considered. The first effect can be with confidence ruled out as a factor limiting the transfer process because decrease of the PL rise time is not consistent with decrease of diffusion coefficient with increasing temperature. The pure carrier capture of carriers situated within the capture region (usually assumed as a distance into which a wave function of a confined carrier is spread out [7]) should show no larger temperature or excitation intensity for the WL, if the WL could be considered as an ordinary quantum well [7]. However, a WL in a self-assembled QD structure exhibits strong strain fields, which induce potential barriers at the interfaces [8]. We believe that it is these barriers that hinder carrier transfer from the GaAs barriers into the WL. Besides, similar barriers due to strain and variations of the InGaAs alloy composition are to be expected between the GaAs layers and the QDs [1].

These potential barriers are most difficult to overcome at low temperatures and for low carrier densities. Increase of the mean carrier energy by increased sample temperature or excitation intensity aids faster carrier transfer over the barriers. On the other hand, with increasing quantum dot density, the average interdot distance decreases (down to 10 nm for the sample E), and the potential barriers overlap, effectively diminishing their influence on the carrier transfer.

For the lowest-density QD structure A, the QD PL rise times are larger that that for the WL. This implies for low QD density structures part of the carriers enter the QDs through the WL, and lateral carrier transport in the WL affects the carrier transfer time, too.

In conclusion, we have performed time-resolved photoluminescence measurements in QD structures with QD densities varying nearly over two orders of magnitude. We have found that the rate of carrier transfer into the dots increases with increased quantum dot density. Our experimental results suggest that at low QD densities carrier transfer is inhibited by potential barriers around the dots and the wetting layer.

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Figure captions

Fig. 1. Time-resolved PL spectra at 100, 840 and 1670 ps after excitation at 78 K for the structures B and D with the dot densities of 7.0×10^{8} and 7.3×10^{9} cm^{-2}, respectively.

Fig. 2. Excitation intensity dependence of the PL rise time at 78 K for the structures A – E and the wetting layers in the low QD density samples. The QD density increases from the sample A to E. The lines are drawn to guide an eye.

Fig. 3. Temperature dependence of the PL rise time for the structures B and E with the QD densities of 7.0×10^{8} and 2.4×10^{10} cm^{-2}.

Fig. 1

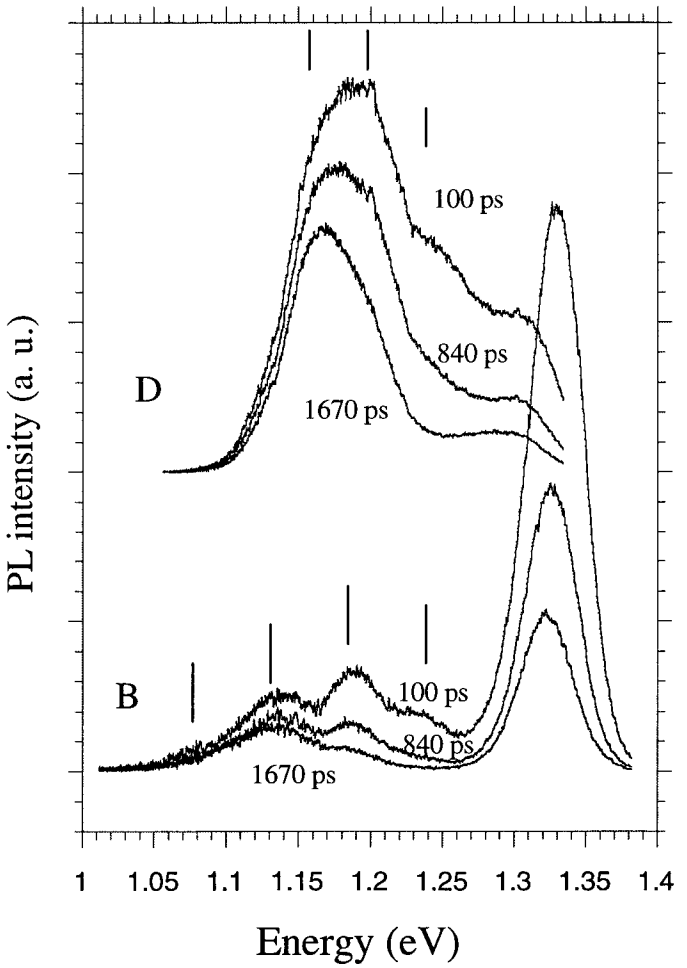

PL intensity (a. u.)
D
100 ps
840 ps
1670 ps
B
100 ps
840 ps
1670 ps
1
1.05
1.1
1.15
1.2
1.25
1.3
1.35
1.4
Energy (eV)

Fig. 2

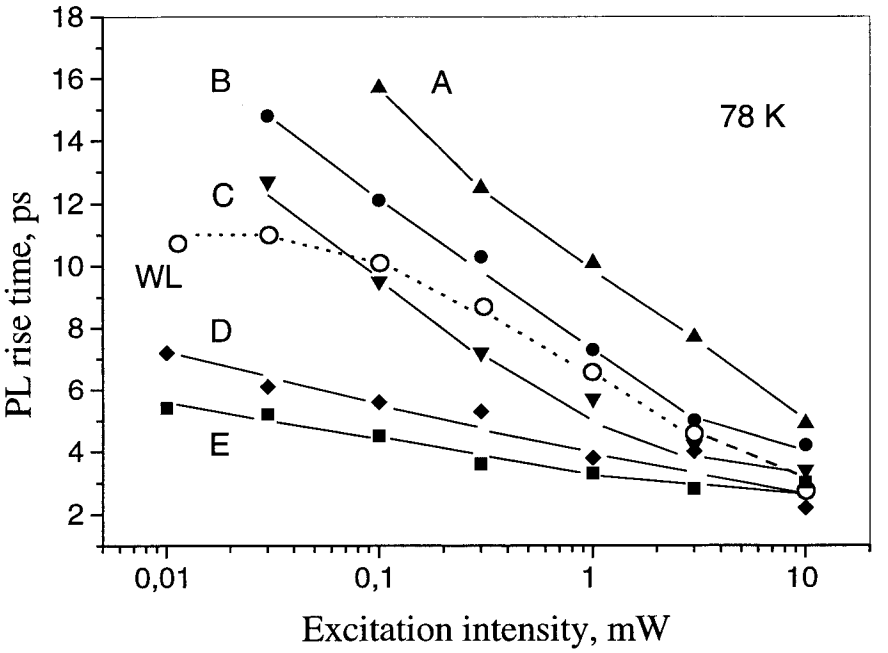
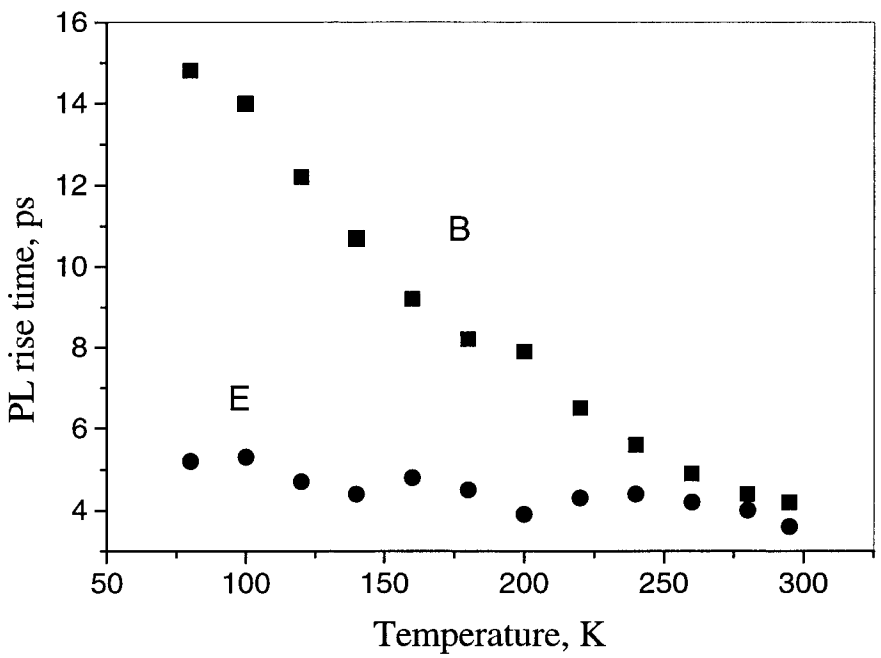

18
16
14
12
10
8
6
4
2
PL rise time, ps
B
A
78 K
C
WL
D
E
0,01
0,1
1
10
Excitation intensity, mW

Fig. 3


16
14
12
10
8
6
4
PL rise time, ps
B
E
50
100
150
200
250
300
Temperature, K